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THE SUPRA-COMPRESSION OF LX-07, LX-17, PBX-9404, and RX-26-AF AND THE EQUATIONS OF STATE OF THE DETONATION PRODUCTS

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Shock velocity vs. input pressure in the range of 32 to 112 GPa has been measured for LX-07, LX-17, PBX-9404, and RX-26-AF. The average shock velocity for the first 2.8-mm of run was found to be within one percent of that for the run from 2.0 to 4.8-mm. Supra-compressive states of PBX-9404 were reflected to higher pressures by a copper barrier and the velocity in the copper measured. They were also rarefied by a magnesium barrier and the velocity in the magnesium measured. Equations of state derived for the reaction products of these explosives and normalized to accepted C-J pressures do not adequately predict measurements at pressures substantially greater than detonation pressure, normalization to lower than accepted C-J pressures is required to fit the data. The C-J state defined by the supra-compressive experiments appears to be different from the C-J state defined by the usual C-J pressure measurement.

INTRODUCTION

A principal purpose of our study was to determine the equation of state (EOS) of HMX and TATB based explosives at pressures well above the Chapman-Jouguet (C-J) pressure.

We have made measurements of the shock velocity for the explosives LX-07, LX-17, PBX-9404, and RX-26-AF, at input pressures of 30 to 110 GPa using our two-stage light gas gun facility.(1) Our data on PBX-9404 agrees with the previous work of Kineke and West(2) allowing for the larger uncertainty in their experimental measurements. In analyzing our results we have found that equations of state that have been derived for the reaction products of these explosives at pressures equal to and less than detonation pressure do not adequately predict measurements for pressures substantially greater than detonation pressure. At present, it appears that the C-J theory may have to be modified somewhat in order to fit all of the data, but this has not yet been proven. There is some indication from our work that the supra-compressive experiments define a different C-J state than that defined by ordinary self-supporting detonations.

EXPERIMENTAL

The supra-compression experiments that are being reported were conducted at LLNL on the two-stage light gas gun using ultrafast pin techniques(1) to monitor the progress of shock waves. Typical experiments consisted of an impactor of either 1100 aluminum or tantalum propelled by the two-stage gun, a buffer plate of either 1100 aluminum or explosive from which time zero is taken, and the specimen of explosive against the buffer plate through which the shock velocity is timed. See Fig. 1. Dimensions were chosen such that the measurements were unaffected by rarefactions. The nominal compositions of the test explosives are given in Table 1. Additional experiments were performed with PBX-9404 where a supra-compressive state was reflected to a much higher pressure by a copper barrier and the shock velocity in the copper measured. In a similar way, a supra-compressive state of PBX-9404 was rarefied by unloading into a magnesium barrier and the shock velocity in the magnesium measured. See Fig. 2. For all experiments, the impactor velocity was determined to better than .1% using flash x-ray techniques. Densities of test explosive, buffer plates, and impactors were determined for the materials used in each experiment. All parts were lapped, polished, and carefully measured to minimize dimensional errors, which could amount to about 2 μm for most materials.

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CALCULATIONS

The ignition and growth model[3] of the reactive flows created during the shock initiation and detonation wave propagation of heterogeneous solid explosives has successfully calculated a great deal of one- and two-dimensional experimental data[4-10]. In this paper the model is applied to the supra-compression data to determine if the effect of the reaction zone is truly negligible, for HMX-based explosive as had been assumed, and to model the LX-17 data, for which the reaction zone does have an effect. The parameters used for PBX-9404 and LX-17 are given in the recent paper of Tarver et al.[11]. Very fine zoning (100 zones per millimeter) is required to accurately describe these small scale, subnanosecond timing experiments. Reactive flow calculations for PBX-9404 do indeed show that the reaction zone effect is negligible in the single and reflected shock experiments shown in fig. 8, because these calculations agreed very closely with C-J burn calculations. In the rarefaction experiments shown in Fig. 9, the reactive flow calculations indicated that the effect of the von Neumann spike decayed in the magnesium plate to a negligible level in just less than the one millimeter thickness, at which the magnesium shock velocity measurements were begun. However, the calculated pressures in the magnesium were always higher than measured, which suggests that the calculations may not have been completely free of reaction zone effects.

Previously reported C-J burn calculations[12] which showed that a P_{CJ} =34.0 GPa equation of state provides a better fit to the supra-compression data than the usual P_{CJ} 37.0 GPa for PBX-9404 have been verified by detailed reactive flow calculations. However, it has been demonstrated that reactive flow calculations using the P_{CJ} = 37.0 GPa reaction products equation of state for PBX-9404 accurately calculate embedded gauge and metal acceleration experiments on self-sustaining detonation waves.[8,9]. Similar calculations with a P_{CJ} = 34.0 GPa equation of state and the usual thin reaction zone for PBX-9404 greatly underestimate the resulting pressures, particle velocities, and metal acceleration properties. Tarver and Urtiew[8] demonstrated that the P_{CJ} = 37.0 GPa equation of state accurately calculates the self-sustaining, converging detonation velocity measurements of Cheret et al.[13]. Since the P_{CJ} = 34.0 GPa equation of state yields higher shock velocities and pressures than the P_{CJ} = 37.0 GPa equation of state, a calculation of Cheret et al.'s data with the P_{CJ} = 34.0 GPa equation of state predicts converging detonation velocities that are much greater than the experimental measurements. Therefore a dilemma exists in that the P_{CJ} = 37.0 GPa reaction product equation of state accurately describes experiments involving self-sustaining detonation waves which are followed by Taylor wave expansions, while the P_{CJ} = 34.0 GPa equation of state matches the supra-compression data and seems to agree more closely with thermodynamic chemical equilibrium code calculations.[14]

The situation is even more complex for TATB-like explosives which produce relatively large amounts of carbon and whose reaction zone lengths are on the order of millimeters thick. Supra-compression experiments on such explosives must be calculated with reactive flow models. The LX-17 supra-compression experiments shown in Fig. 4 were calculated using the parameters given by Tarver et al.[11] In the lowest pressure experiment, the calculated higher average shock velocity in the 2-4 mm depth agreed very closely with the experimental observation. In the higher pressure experiments, the reactive flow calculations yield constant average shock velocities with run distance, although the shape of the reaction zone changes slightly during each calculation. Both the cited[15] P_{CJ} = 30.0 GPa and also a highly modified JWL reaction product equation of state with P_{CJ} = 27.5 GPa and D = 7.64 km/s yield calculated supra-compressive shock velocities that are lower than the experimental values at the higher pressures. A lower P_{CJ} value (~25.GPa) allows a much better match between calculations and supra-compressive experiments (but not with P_{CJ} measurements).

EXPERIMENTAL RESULTS

The single-shock supra-compressive experiments on LX-07, LX-17, PBX-9404, and RX-26-AF are represented schematically in Fig. 1 and summarized in Table 2 and in Figs. 3 through 7. Curves from the best JWL equations of state from cylinder test data[15] normalized to accepted C-J pressures are shown in the same figures. According to Chapman/Jouguet theory, the slope of the Hugoniot shock velocity vs. particle velocity detonation product curve should be zero at the C-J point, and the locus of pressure vs. relative volume states should be tangent to the Rayleigh line through the C-J point at the C-J point. An examination of Figs. 3 through 8 shows the difficulty. The supra-compressive data does not appear to be consistent with C-J theory in that the Hugoniot shock velocity vs. particle velocity curves do not appear to go to zero slope at the C-J points and the locus of pressure vs. relative volume states do not appear to become tangent to the Rayleigh lines at the accepted C-J points for the explosives tested. The data does not prove that C-J conditions can not be met as one approaches the C-J state from the overdriven side but it strongly suggests that this is the case. If there were a significant change in slope of the shock velocity vs. particle velocity curve very near to the C-J state, it would not be detected by the present technique. The error limits are simply too large to permit resolution.

The reflected-shock supra compressive experiments on PBX-9404 are summarized in Table

3 and in Fig. 8. A suitable choice of parameters would allow a reactive-flow hydrodynamic calculation of both the single-shock and reflected-shock experiments within experimental error. However, the same parameters do not accurately predict the initial motion of a thin metal plate being accelerated by a head on detonation of PBX-9404 in a metal acceleration experiment, the metal moving faster than predicted. The definition of C-J state appears to be different for the supra-compressive experiments.

The experiments on rarefaction of supra compressive states of PBX-9404 are summarized in Table 3 and in Fig. 9. Differences between equation of state assumptions (i.e., P_{CJ} = 34 GPa vs. P_{CJ} = 37 GPa) are too small to resolve the question of which describes the data better. The best value appears to lie between these two pressures. The experimental values of pressure are lower than calculated, probably due to reaction zone effects in the calculation. The error limits in both calculation and experiment are such that an accurate determination of the point of tangency to the Rayleigh line cannot be made within the accuracy required to determine the "C-J" pressure.

DISCUSSION

It can be reasonably postulated that the C-J detonation pressure of LX-07, LX-17, PBX-9404, and RX-26-AF as implied by supra compression experiments really is lower than that measured in the usual detonation pressure measurements. The 34.0 GPa JWL calculation shown in Fig. 5 for PBX-9404 illustrates that a calculation based on a lower P_{CJ} can be fit to the supra-compressive data and have zero slope at the lower P_{CJ}. Such fits based on lower P_{CJ} can be made to the other data as well.(2) The question then becomes "why is there a difference between P_{CJ} as usually measured and P_{CJ} as defined in a supra-compressive experiment?" It is suggested that this lower lower P_{CJ} could occur if a relatively slow reaction (for example, carbon condensation or diffusion controlled equilibration of reaction products from different components of the explosive) followed a relatively fast reaction. With the usual measurements, any changes from a second, relatively slow reaction during expansion of products from the more rapid first reactions(s)) would be extremely difficult to measure. Careful measurements can detect only the end of the relatively fast reactions(s)), which is taken to be the C-J state. This state has most of the attributes of the C-J state and for many purposes may be assumed to be the C-J state. However, if the postulate just made is true, it is not an equilibrium state and hence, not a C-J state, and would be subject to scale effects to some degree.

Whether or not detonation velocity, for example, would be measurably affected by slow reactions in large charges is a matter of conjecture. Careful pressure or particle velocity vs. time measurements should be able to detect the effects of any slow reactions in large charges.

Supra compressive experiments on LX-17 offer an interesting contrast with the other explosives tested. Because carbon is present in the detonation products of LX-17 to a much greater extent than in the products of PBX-9404, if carbon condensation were the "slow" reaction responsible for the differences in apparent P_{CJ}, then LX-17 might be expected to show an even larger difference between measured C-J detonation pressure and that inferred from supra compression experiments. Such appears to be the case. It should be noted that this is true in spite of the fact that the reaction zone as usually measured is much longer for TATB-based explosives than for HMX-based explosives, so there is much more time for "slower" reactions to take place, which should tend to offset differences. A single equation of state of the detonation products of LX-17 is not adequate to describe expanded states, the C-J state, and supra compression states when incorporated into reactive flow hydrodynamic calculations. TATB and HMX-based systems appear similar in this respect.

An accurate description of both expansion and supra-compressive reaction product states with a single equation of state may not be possible if P_{CJ} as normally measured is in fact not an equilibrium state. A lack of equilibrium would imply chemical differences between the C-J states defined by the usual methods and that inferred from supra-compressive experiments. The discontinuities seen in derivative functions near the C-J point[12,16,17] as one moves from expansion states to supra-compressive states could be due to such chemical differences.

Experiments on PETN and other relatively oxygen rich explosives where there is very little carbon in the detonation products should help resolve these questions arising from the experiments on HMX and TATB-based explosives. A reactive flow model consisting of a fast exothermic reaction to one reaction product equation of state followed by a slower one to a lower C-J pressure reaction product equation of state had been proposed to explore the thermodynamic and reaction time parameters involved in this phenomenon. The current explanation centers on carbon condensation due to its thermodynamic properties and the chemical equilibrium calculations.[14]

Table I. Compositions of Test Explosives

Explosives	Wt.% Composition
LX-07	90/10 HMX/Viton A
LX-17	92.5/7.5 TATB/Kel-F 800
PBX-9404	94/3/3 HMX/NC/CEP
RX-26-AF	45/48.5/6.5 TATB/HMX/Estane

Table II. Conditions and Results of Single-Shock Supra-Compressive Experiments.

Experi-ment	Impactor Velocity km/s	Impactor Material	Impactor Density g/cm³	Buffer Plate Material	Buffer Plate Density g/cm³	Target Material	Target Density	Target Shock Vel. km/s±2σ	Target Particle Vel. km/s±2σ	Target Pressure GPa	Target Rel. Vol. V/V_o	Target Run Measured mm
MG 8	4.796	1100 Al	2.712	1100Al	2.716	LX-07	1.86[illegible]	9.00 ±.06	2.730 ±.020	45.72	.6967	0 to 2.8
MG 24	4.892	1100 Al	2.707	LX-07	1.858	LX-07	1.860	8.94 ±.03	2.796 ±.007	46.49	.6872	2 to 4.8
MG 9	5.961	1100 Al	2.714	1100 Al	2.720	LX-07	1.86[illegible]	9.57 ±.06	3.410 ±.029	60.80	.6437	0 to 2.8
MG 10	7.544	1100 Al	2.715	1100 Al	2.721	LX-07	1.864	10.33 ±.07	4.367 ±.030	93.90	.5771	0 to 2.8
MG 29	3.508	1100 Al	2.713	1100 Al	2.704	LX-17	1.907	7.44 ±.05				0 to 2.0
						LX-17	1.90[illegible]	7.68 ±.06				2 to 4.0
MG 30	3.813	1100 Al	2.713	1100 Al	2.711	LX-17	1.906	7.70 ±.04	2.222 ±.015	32.61	.7114	0 to 2.0
						LX-17	1.90[illegible]	7.70 ±.04	2.223 ±.015	35.57	.7113	2 to 4.0
MG 32	4.400	1100 Al	2.712	LX-17	1.899	LX-17	1.901	8.02 ±.04	2.563 ±.013	39.08	.6804	2 to 4.0
						LX-17	1.898	7.98 ±.04	2.569 ±.013	38.91	.6780	4 to 6.0
MG 36	4.436	1100 Al	2.712	LX-17	1.905	LX-17	1.905	8.00 ±.02	2.587 ±.007	39.42	.6766	2 to 4.8
MG 35	5.355	1100 Al	2.712	LX-17	1.903	LX-17	1.903	8.44 ±.04	3.134 ±.015	50.34	.6286	2 to 4.8
MG 31	5.612	1100 Al	2.713	1100 Al	2.705	LX-17	1.90[illegible]	8.60 ±.0[illegible]	3.284 ±.014	53.72	.6181	0 to 4.0
MG 13	4.164	1100 Al	2.714	PBX-9404	1.844	PBX-9404	1.844	8.85 ±.04	3.353 ±.012	38.40	.7341	2 to 6.9
MG 4	4.779	1100 Al	2.714	1100 Al	2.719	PBX-9404	1.841	9.07 ±.06	2.723 ±.020	45.47	.6998	0 to 2.8
MG 3	5.032	1100 Al	2.715	1100 Al	2.717	PBX-9404	1.841	9.17 ±.08	2.874 ±.024	48.52	.6866	0 to 2.8
MG 16	5.901	1100 Al	2.715	PBX-9404	1.845	PBX-9404	1.844	9.47 ±.05	3.399 ±.017	59.35	.6411	2 to 4.8
MG 15	6.218	1100 Al	2.715	PBX-9404	1.846	PBX-9404	1.844	9.59 ±.10	3.591 ±.036	63.50	.6256	2 to 4.8
MG 1	7.533	1100 Al	2.715	1100 Al	2.718	PBX-9404	1.84[illegible]	10.3[illegible] ±.07	4.343 ±.020	82.92	.5811	0 to 2.8
MG 14	6.402	Ta	16.675	PBX-9404	1.846	PBX-9404	1.84	11.0[illegible] ±.06	5.130 ±.031	105.14	.5380	2 to 4.8
MG 12	6.760	Ta	16.681	PBX-9404	1.845	PBX-9404	1.84[illegible]	[illegible]	5.424 ±.036	112.27	.5170	2 to 4.8
MG 20	4.547	1100 Al	2.715	1100 Al	2.711	RX-26-AF	1.8[illegible]	8.50 ±.04	2.641 ±.01[illegible]	41.20	.6893	0 to 2.8
MG 5	4.710	1100 Al	2.712	1100 Al	2.713	RX-26-AF	1.8[illegible]	8.62 ±.08	2.730 ±.026	43.24	.6832	0 to 2.8
MG 19	5.028	1100 Al	2.716	1100 Al	2.716	RX-26-AF	1.83[illegible]	8.72 ±.02	2.929 ±.01[illegible]	46.79	.6641	0 to 2.8
MG 25	5.416	1100 Al	2.715	RX-26-AF	1.835	RX-26-AF	1.836	8.82 ±.04	3.167 ±.015	51.28	.6410	2 to 4.8
MG 6	5.988	1100 Al	2.714	1100 Al	2.713	RX-26-AF	1.8[illegible]	9.14 ±.08	3.500 ±.035	58.77	.6171	0 to 2.8
MG 7	7.493	1100 Al	2.715	1100 Al	2.716	RX-26-AF	1.8[illegible]	10.0[illegible] ±.0[illegible]	4.361 ±.03[illegible]	80.83	.5678	0 to 2.8

Table III. Conditions and Results of Reflection and Rarefaction Experiments on Supra Compressive States of PBX-9404. See Fig. 2 for Experimental Configuration

Experiment	Impactor Velocity km/s	Impactor Material	Impactor Density g/cm³	PBX-9404 Density g/cm³	Impact Pressure* PBX-9404 GPa	Barrier Material	Barrier Density g/cm³	Barrier Shock Velocity km/s±2σ	Barrier Pressure GPa
MG 18	4.310	1100 Al	2.715	1.841	40.[illegible]	Cu	8.93	5.864±.026	66.41
MG 17	6.055	1100 Al	2.715	1.838	61.[illegible]	Cu	8.93	6.67±.10	107.25
MG 23	6.294	1000 Al	2.713	1.840	64.6[illegible]	Cu	8.93	6.759 ±.022	112.22
MG 27	3.996	1100 Al	2.716	1.840	36.[illegible]	Mg	1.738	7.55 ±.06	32.22
MG 26	4.200	1100 Al	2.706	1.840	38.[illegible]	Mg	1.736	7.70 ±.07	34.50
MG 28	4.432	1100 Al	2.714	1.840	41.[illegible]	Mg	1.7[illegible]3	7.82 ±.06	36.45

* Calculated from best fit to single-shock supra-compressive data.

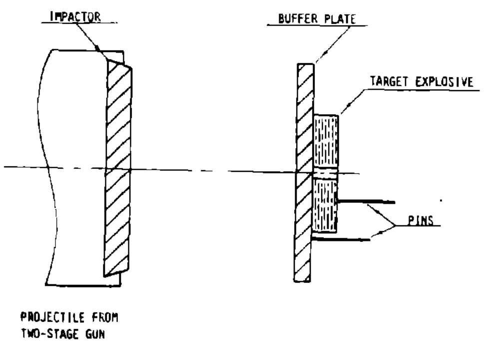


Fig. 1 Test configuration for single-shock supra-compression experiments on LX-07, LX-17, PBX-9404 and RX-26-AF. Some of the test samples of LX-17 were two-level and additional pins were used to enable accurate velocity stability determinations to be made.

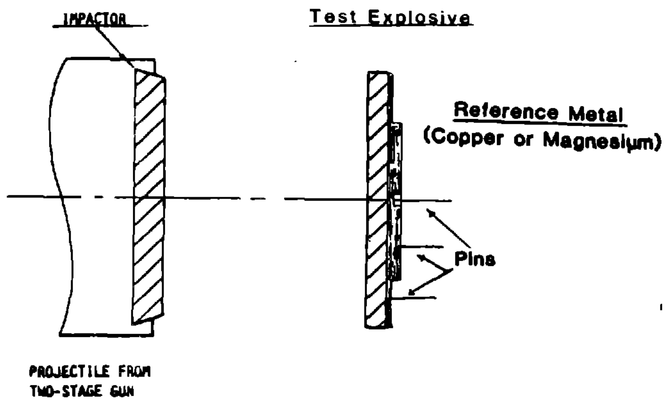


Fig. 2 Test configuration for reshock and rarefaction of supra-compressive states of PBX-9404.

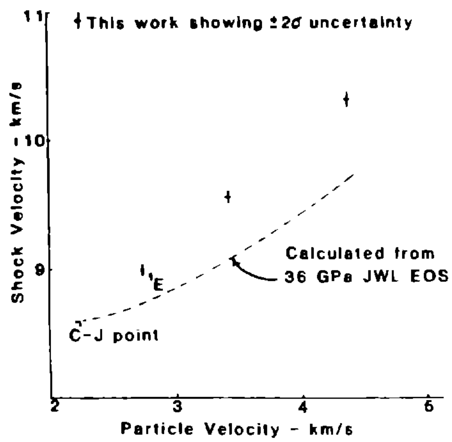


Fig. 3 Experimental LX-07 supra-compressive data. Measured shock velocity is vs. particle velocity derived from flyer velocity, density, and Hugoniot relationships. Data points marked "E" used an LX-07 buffer plate. All others used an 1100 Al buffer plate. See Fig. 1.

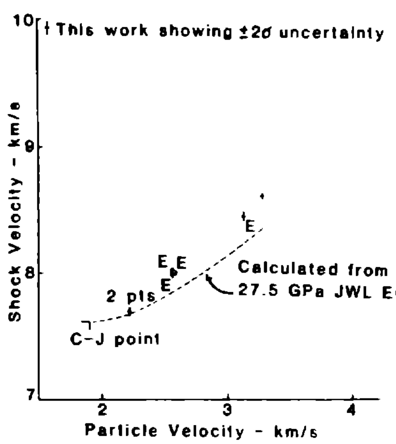


Fig. 4 Experimental LX-17 supra-compressive data. Measured shock velocity is vs. particle velocity derived from flyer velocity, density, and Hugoniot relationships. Data points marked "E" used an LX-17 buffer plate. All others used an 1100 Al buffer plate. See Fig. 1.

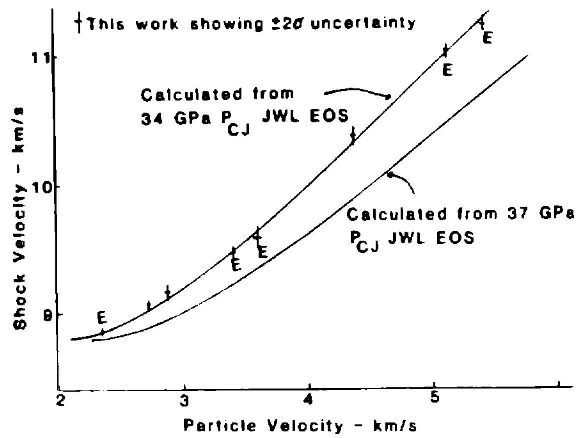


Fig. 5 Experimental PBX-9404 supra-compressive data. Measured shock velocity is vs. particle velocity derived from flyer velocity, density, and Hugoniot relationships. Data points marked "E" used an PBX-9404 buffer plate. All others used an 1100 Al buffer plate. See Fig. 1.

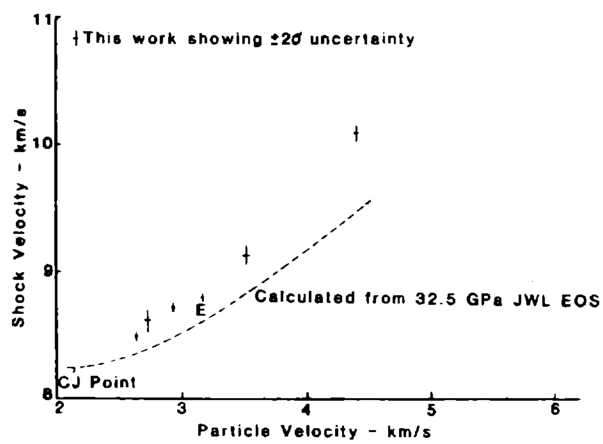


Fig. 6 Experimental RX-26-AF supra-compressive data. Measured shock velocity is vs. particle velocity derived from flyer velocity, density, and Hugoniot relationships. Data points marked "E" used an RX-26-AF buffer plate. All others used an 1100 Al buffer plate. See Fig. 1.

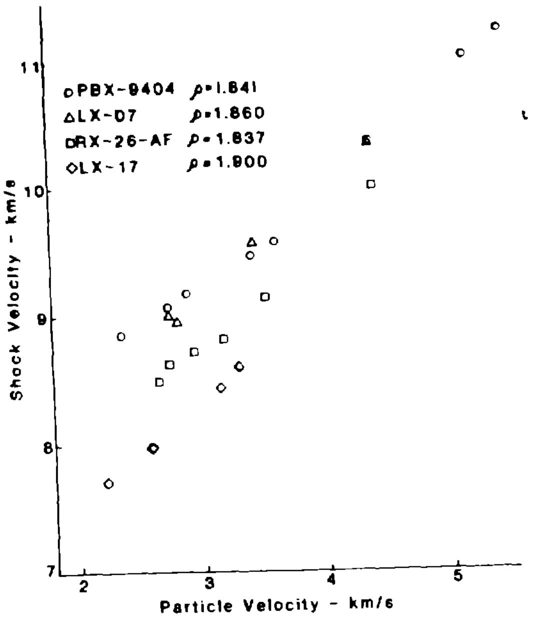
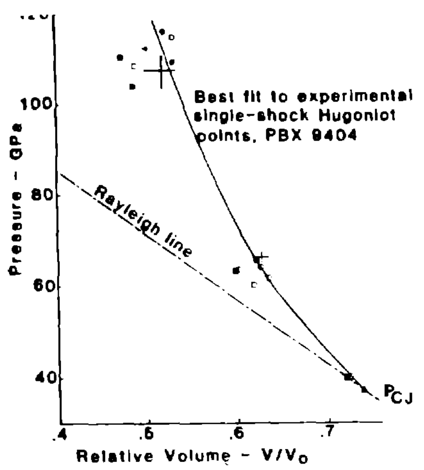


Fig. 7 Combined plot of shock velocity vs. particle velocity for all single-shock supra-compressive experiments to illustrate systematics of data.



○ Calculated for single shock using 34.0 GPa P_{CJ} JWL EOS
● Calculated for reflected shock off of copper using 34.0 GPa P_{CJ} JWL EOS.
□ Calculated for single shock using published 37.0 GPa P_{CJ} JWL EOS.
■ Calculated for reflected shock off of copper using published 37.0 GPa P_{CJ} JWL EOS.
† Experimental reflected shock values off of copper, ±2σ.

Fig. 8 Reflected and single-shock supra-compressive states of PBX-9404. Calculational results using two different equations of state are compared with experiment.

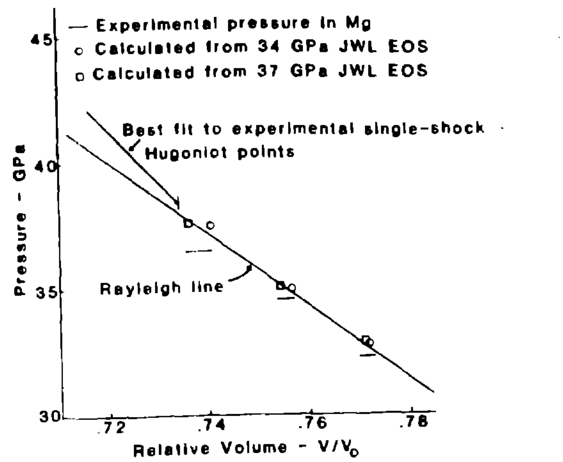


Fig. 9 Insentrope states from PBX-9404/Mg experiments near P_{CJ}. Calculations of pressure vs. relative volume for individual experiments using two different JWL equations of state are compared with experimental pressures calculated from shock velocity measured in magnesium using the same shock Hugoniot ($U_S = 4.52 + 1.233\ U_P$) for the calculations and the shock velocity to pressure conversions.

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